

AFRL-AFOSR-UK-TR-2012-0006



**Novel, High-power, Mid-infrared Optical Source
for the 5-12 Micron Spectrum**

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EOARD GRANT 09-3017

Report Date: December 2011

Final Report for 20 November 2008 to 20 November 2011

Distribution Statement A: Approved for public release distribution is unlimited.

**Air Force Research Laboratory
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REPORT DOCUMENTATION PAGE				Form Approved OMB No. 0704-0188	
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1. REPORT DATE (DD-MM-YYYY) 05-03-2012		2. REPORT TYPE Final Report		3. DATES COVERED (From – To) 20 November 2008 – 20 November 2011	
4. TITLE AND SUBTITLE Novel, High-power, Mid-infrared Optical Source for the 5-12 Micron Spectrum				5a. CONTRACT NUMBER FA8655-09-1-3017	
				5b. GRANT NUMBER Grant 09-3017	
				5c. PROGRAM ELEMENT NUMBER 61102F	
				5d. PROJECT NUMBER	
6. AUTHOR(S) Professor Majid Ebrahim-Zadeh				5d. TASK NUMBER	
				5e. WORK UNIT NUMBER	
7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES) ICFO - Institut de Ciencies Fotoniques (ICFO - The Institute of Photonic Sciences) Mediterranean Technology Park, Castelldefels Barcelona, Spain 08860				8. PERFORMING ORGANIZATION REPORT NUMBER N/A	
9. SPONSORING/MONITORING AGENCY NAME(S) AND ADDRESS(ES) EOARD Unit 4515 BOX 14 APO AE 09421				10. SPONSOR/MONITOR'S ACRONYM(S) AFRL/AFOSR/RSW (EOARD)	
				11. SPONSOR/MONITOR'S REPORT NUMBER(S) AFRL-AFOSR-UK-TR-2012-0006	
12. DISTRIBUTION/AVAILABILITY STATEMENT Approved for public release; distribution is unlimited.					
13. SUPPLEMENTARY NOTES					
14. ABSTRACT This project has led to important new advances in coherent near- to mid-infrared light sources on several fronts. By exploiting nonlinear frequency down-conversion techniques based on optical parametric generation and oscillation, we have developed several novel optical sources in different time-scales and operating regimes with extended tuning throughout the 1.4-4.2 μm spectral range in the near- to mid-IR and 6-6.8 μm in the deep mid-infrared. The developed devices exploit the new generation of QPM and mid-IR nonlinear crystals of MgO:PPLN, MgO:sPPLT, and CdSiP ₂ together with fiber and solid-state laser technology at 1064 nm as the pump source, resulting in compact, practical, and portable device architectures for many real applications. We have achieved record optical powers of >17 W in cw operation and average powers up to 12 W in the ultrafast picosecond regime at 80 MHz high-repetition-rate, covering the spectral range of 1.4-4.2 μm . In the deep mid-IR, we have achieved record pulse energies as much as 1.5 mJ at repetition rates as high as 450 MHz, with spectral coverage across 6-6.8 μm . The developed systems have provided a viable new class of tunable coherent light sources for previously inaccessible spectral regions in the near- to mid-infrared offering record optical powers and pulse energies. With the exploitation of mid-IR optical crystals of ZnGeP ₂ and OPGaAs of high optical quality in cascaded pumping schemes using the near- to mid-IR parametric sources developed during this project, further wavelength extension into the 5-12 μm spectral range will be feasible. The exploitation of newly emerging Tm fiber laser technology near 2 μm also offers great potential for direct generation of tunable coherent radiation at unprecedented optical powers in the 5-12 μm spectral range using one-step parametric generation in the nonlinear crystals of ZnGeP ₂ , CdSiP ₂ , and OP-GaAs. These exciting new avenues for further research and development are currently underway.					
15. SUBJECT TERMS EOARD, Non-linear Optics, Optical parametric oscillators					
16. SECURITY CLASSIFICATION OF:			17. LIMITATION OF ABSTRACT SAR	18. NUMBER OF PAGES 24	19a. NAME OF RESPONSIBLE PERSON A. GAVRIELIDES
a. REPORT UNCLAS	b. ABSTRACT UNCLAS	c. THIS PAGE UNCLAS			19b. TELEPHONE NUMBER (Include area code) +44 (0)1895 616205

Proposal Title:

**Novel, high-power, mid-IR optical source for the 5-12 μm spectrum
(EOARD Grant: FA8655-09-1-3017; 20 Nov 2008 - 30 Nov 2011)**

Final Report (10 December 2011)

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Abstract

The goal of this proposal has been the development of a new class of advanced solid-state photonic source for the difficult spectral regions beyond 5 μm in the mid-infrared (mid-IR), where there exists a severe shortage of practical coherent laser sources, and many scientific and technological applications, from trace gas detection and environmental sensing to safety, security, defense, and biomedicine can potentially benefit. The strategy has been to exploit nonlinear optical techniques based on parametric generation and oscillation in the latest class of mid-IR nonlinear materials in combination with the most advanced lasers pump sources based on solid-state and fiber laser technology, to realize novel, high-power coherent source with tunability beyond 5 μm in the mid-IR. During the course of the project, we were successful in achieving several breakthroughs in the development of a number of unique parametric sources for >5 μm spectral range operating in different time scales, providing record pulse energies, average powers, and conversion efficiencies in practical, compact, scalable, and cost-effective designs. Our research efforts during the course of this project has resulted in 9 technical papers in the leading international journals, 3 proceedings publications, 2 major invited book chapters and reviews, 7 peer-reviewed conference contributions and 22 invited talks at major international meetings and workshops.

This final report provides a review of the research achievements during the period of the project, 20 Nov 2008 to 30 Nov 2011. The report details the most recent progress and the new results obtained since the submission of the 5th progress report in May 2011, while summarizing the research results presented in earlier progress reports with the relevant references included.

1. Fiber-laser-pumped, multiwatt, near- to mid-infrared, continuous-wave optical parametric oscillator based on MgO:PPLN

In one particular track of investigation, we devoted considerable effort to the development of continuous-wave (cw) optical parametric oscillators (OPO) for the near- to mid-infrared to provide tunable high-power radiation in the 1.5–4 μm spectral region. The output radiation from such a *primary* OPO would then be used as the pump source for a *secondary* OPO in a *cascaded* pumping scheme to achieve wavelength generation beyond 5 μm . In order to provide the most practical, compact, portable, and cost-effective configuration, with the potential for future power scaling, we deployed cw Yb fiber laser technology at 1064 nm as the primary laser pump source.

In our early effort in this direction, we successfully developed a stable, high-power, fiber-laser-pumped, continuous-wave OPO in singly-resonant oscillator (SRO) configuration using the quasi-phase-matched (QPM) nonlinear material, MgO:PPLN, as the gain medium [1]. Using a single-frequency, cw Yb fiber laser at 1064 nm and a 50-mm-long MgO:PPLN crystal, by optimization of signal output coupling we generated up to 9.8 W of signal together with 7.7 W of idler power for 28.6 W of pump power, resulting in a total output power of 17.5 W at 61% extraction efficiency. We achieved a total tuning range of 513 nm (120 nm of signal over 1594–1714 nm, 393 nm of idler over 2803–3196 nm) across which watt-level output power was extracted. By careful control of thermal effects, we obtained a long-term peak-to-peak idler power stability of 5% over 14 hours near room temperature. The output beams were also characterized by TEM₀₀ spatial profile with $M^2 < 1.28$ for the idler and $M^2 < 1.37$ for the signal, which are important for many practical applications. Detailed discussion of the design parameters and performance characteristics of this OPO has already been presented in interim Progress Reports 1 & 2.

We also developed a compact and viable source of broadband, high-power, mid-infrared radiation based on the same cw MgO:PPLN OPO pumped by a wide-bandwidth cw Yb fiber laser centered at 1060 nm [2]. By exploiting the extended phase-matching bandwidth in a 50 mm crystal of MgO:PPLN crystal and a ring SRO cavity, we obtain 5.3 W of broadband idler output for 25.5 W of input pump power at >80% pump depletion, transferring a pump bandwidth of 8.3 nm to an idler spectrum spread across 76 nm about the central wavelength of 3460 nm. By deploying output coupling of the signal, we generate 11.2 W of total power at 44% extraction efficiency with pump depletions of >73% at the maximum available pump power. Measurements of transverse modal power confirm Gaussian distribution of the signal and idler output beams. Detailed discussion of the design parameters and performance characteristics of this OPO has already been presented in interim Progress Report 3.

2. Fiber-laser-pumped, multiwatt, near- to mid-infrared, continuous-wave optical parametric oscillator based on MgO:sPPLT

More recently, we developed a stable and high-power operation of a cw SRO using the newly emerging QPM nonlinear material, MgO:sPPLT, pumped by the same cw Yb fiber laser at 1064 nm [3]. The SRO, based on a 30-mm-long crystal and configured in a compact ring resonator, was tunable over 430 nm from 3032-3462 nm in the idler, and could generate as much as 5.5 W of mid-IR idler output power, with >4W of over 60% of the tuning range. This performance was achieved under reduced thermal effects, enabling room temperature operation of the device. The idler output at 3299 nm was characterized by a peak-to-peak power stability better than 12.8% over 5 hours and wavelength excursion of ~32 pm (1 GHz), while operating close to room temperature, and exhibited a linewidth of ~0.2 nm. The corresponding signal linewidth at 1570 nm was ~21 MHz. Idler power scaling measurements were also compared with a MgO:PPLN cw SRO developed earlier [1], confirming that MgO:sPPLT is also an attractive material for multiwatt mid-IR generation, particularly with regard to room temperature operation which is of great advantage in many practical applications.

2.1. Experimental design

The configuration of the cw SRO based on MgO:sPPLT was similar to that in our earlier work [1]. The pump source was a cw, single-frequency Yb fiber laser (IPG Photonics, YLR-30-1064-LP-SF), delivering up to 30 W at 1064 nm in a linearly polarized beam of 4 mm diameter in TEM₀₀ spatial mode ($M^2 < 1.01$), with a linewidth of 89 kHz. The MgO:sPPLT crystal was a 30-mm-long, 1-mm-thick sample with six grating periods from $\Lambda = 29.15 \mu\text{m}$ to $30.65 \mu\text{m}$. It was housed in an oven with a temperature stability of $\pm 0.1 \text{ }^\circ\text{C}$. The SRO cavity was a symmetric ring, formed by two concave ($r = 100 \text{ mm}$) and two plane mirrors. All mirrors had high reflectivity ($R > 99\%$) over 1.3-1.9 μm and high transmission ($T > 90\%$) over 2.2-4 μm , ensuring SRO operation. For frequency control, a 500- μm -thick uncoated fused silica etalon (free spectral range, FSR~205 GHz) was used at the second cavity waist between the plane mirrors. The pump beam was confocally focused to a beam radius of 48 μm ($\xi \sim 1$) at the centre of the crystal. The cavity design ensured optimum overlap of pump and resonant signal at the center of the crystal ($b_p \sim b_s$), with a signal waist radius of 58 μm . A dichroic mirror was used to separate the generated output idler from the pump. The total optical length of the cavity including the crystal and the etalon was 575 mm, corresponding to a FSR~522 MHz.

2.2. Tuning characteristics

In order to characterize the cw SRO with regard to tunability, we varied the temperature of the MgO:sPPLT crystal from 40 $^\circ\text{C}$ to 200 $^\circ\text{C}$. The temperature tuning curves for two grating periods,

$\Lambda=30.65 \mu\text{m}$ and $30.15 \mu\text{m}$ are shown in Fig. 1. The signal wavelength was monitored using a near-IR spectrum analyzer, while the idler wavelength was measured using a wavelength meter. The solid lines are the theoretical tuning curves calculated from the relevant Sellmeier equations [4]. From Fig. 1, it can be seen that the experimental measurements are in good agreement with the calculated tuning curves.

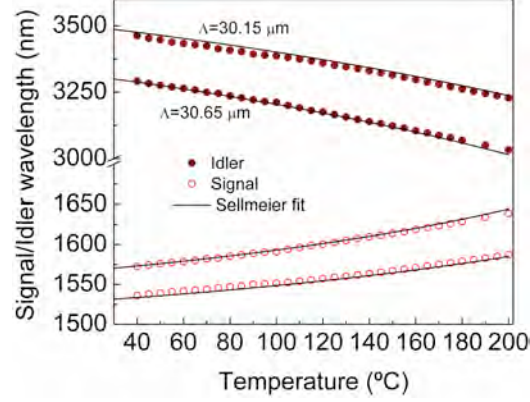


Fig. 1. Temperature tuning curves of 1064 nm-pumped MgO:sPPLT cw SRO.

2.3. Output power

Figure 2(a) shows the idler power extracted from the MgO:sPPLT cw SRO across the mid-IR tuning range. The data were obtained at an available pump power of 28.5 W at the input to the MgO:sPPLT crystal. Using the two grating periods ($\Lambda=30.65, 30.15 \mu\text{m}$), temperature tuning the SRO from room temperature to 200 °C resulted in the generation of idler wavelengths from 3032 to 3462 nm, corresponding to a total tuning range of 430 nm, with a maximum idler power of 5.5 W at 3221 nm and >4 W over more than 60% of the tuning range. The corresponding pump depletion across the tuning range is shown in Fig. 2(b), where it can be seen that pump depletion $>50\%$ is achieved over more than 40% of the tuning range. In addition to the idler, a small amount of signal power (few tens of milliwatts) is also obtained as leakage through the plane mirrors of the ring cavity over the entire tuning range. It is interesting to note that although the thermal load in the nonlinear crystal due to pump, idler and the high intracavity signal cannot be completely ignored, unlike in MgO:PPLN cw SRO [1], high-power operation of the MgO:sPPLT cw SRO can be easily achieved at lower temperatures down to ~ 35 °C. In contrast, operation of MgO:PPLN cw SRO at high power is not attainable below ~ 45 °C due to increased thermal effects [1]. The reduced thermal effects in MgO:sPPLT cw SRO can be attributed to intrinsic material properties including higher thermal conductivity (8.4 W/m-K), better transmission, and lower circulating intracavity signal power due to lower d_{eff} as compared to MgO:PPLN. Also, the shorter interaction length (30 mm) of the nonlinear crystal results in reduced absorption at pump, signal and particularly the idler, which become significant at longer wavelengths.

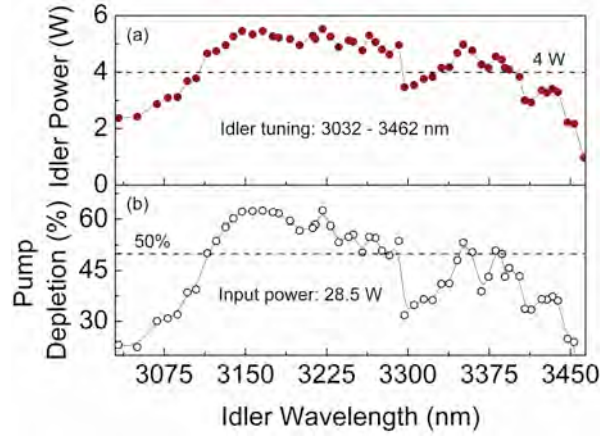


Fig. 2. (a) Idler power, and (b) Pump depletion across the tuning range of MgO:sPPLT cw SRO.

2. 4. Power scaling

We performed the idler power scaling measurements at different wavelengths across the mid-IR tuning range. Fig. 3(a) shows the results obtained at 40 °C for a grating period of $\Lambda=30.65 \mu\text{m}$ (idler wavelength of 3291 nm). Also included for comparison is the power scaling results for a cw SRO based on a 50-mm-long MgO:PPLN at a similar wavelength and operating under the same conditions [1].

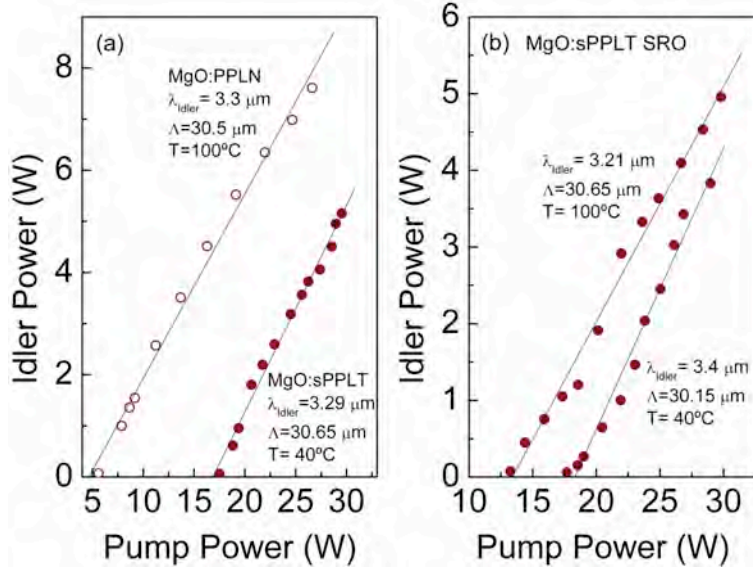


Fig. 3. (a) Idler power scaling comparison of MgO:PPLN and MgO:sPPLT cw SROs at $\sim 3.3 \mu\text{m}$,
(b) Idler power scaling of MgO:sPPLT cw SRO at $3.21 \mu\text{m}$ and $3.4 \mu\text{m}$.

Owing to the lower d_{eff} and shorter crystal length (30 mm), the threshold pump power of the MgO:sPPLT cw SRO is recorded to be 17.5 W, while that of the MgO:PPLN cw SRO is only 5.6 W. However, a maximum idler power of 5.2 W is generated for a pump power of 28.1 W at an idler efficiency of 18.5%, with no saturation in the idler power observed. The corresponding maximum idler power generated in the MgO:PPLN cw SRO is 7.6 W for 26.6 W of pump power at an idler efficiency of 28.6%. On the other hand, we also characterized the SRO at a higher temperature of 100 °C using the same grating period,

corresponding to an idler wavelength of 3212 nm, where we obtained a reduced pump power threshold of 13.2 W and generated as much as 5 W of idler for 29.8 W of pump power, as shown in Fig. 3(b). Also shown in Fig. 3(b) is the power scaling at an idler wavelength of 3403 nm, generating 3.8 W of idler for a pump power of 29 W, at temperature of 40 °C using a grating period of $\Lambda=30.15$ μm . These measurements confirm that despite the lower d_{eff} of the material compared to MgO:PPLN, resulting in higher pump threshold, multiwatt idler output powers in the mid-IR can be generated in cw SROs using MgO:sPPLT.

2. 5. Power and wavelength stability

Further, we recorded the idler power stability of the MgO:sPPLT cw SRO close to room temperature at ~ 35 °C (with the oven switched off), corresponding to an idler wavelength of 3299 nm. We obtained a peak-to-peak power stability better than 12.8% over 5 hours at an idler power >4.5 W, as shown in Fig. 4. The corresponding signal spectrum at 1570 nm, obtained with the intracavity etalon, and recorded using a Fabry-perot interferometer (FSR=1 GHz, finesse=400), is also shown in the inset of Fig. 4, confirming single-frequency operation with an instantaneous linewidth of ~ 21 MHz.

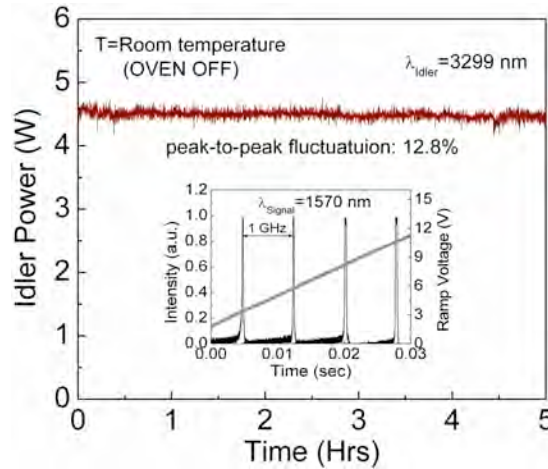


Fig. 4. Idler power stability over 5 hours at room temperature, and corresponding signal single-frequency spectrum (inset).

Under similar conditions, we also recorded the idler wavelength stability using a wavemeter (Bristol 721B-IR) with an absolute accuracy of 1 ppm and a measurement rate of ~ 0.7 Hz. Figure 5 shows the idler wavelength stability recorded over a period of 1 hour, confirming a peak-to-peak wavelength excursion of ~ 32 pm (1 GHz) without any active stabilization. With better thermal isolation and active control, further improvement in the SRO wavelength stability is expected. Also shown in the inset of Fig. 5 is the measured idler spectrum centered at 3299 nm with an FWHM linewidth ~ 0.2 nm, limited by the resolution of the wavemeter. Similar linewidths have been measured at other signal and idler wavelengths across the tuning range. Given the single-frequency nature of the Yb fiber laser pump source with a typical linewidth of 89 KHz, we expect the generated idler wave from the SRO also to be in a single axial

mode. However, more precise measurement of idler linewidth requires other methods such as beat frequency technique.

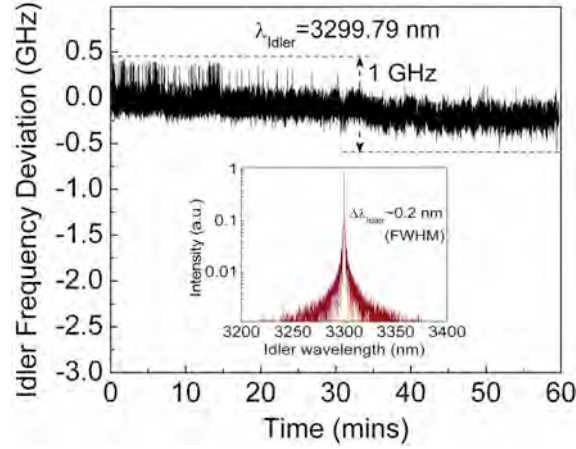


Fig. 5. Idler wavelength stability over 1 hour at room temperature and corresponding idler spectrum (inset).

3. Fiber-laser-pumped, multiwatt, near- to mid-infrared, synchronously-pumped picosecond optical parametric oscillator based on MgO:PPLN

In a parallel research effort, we embarked on the development of new OPO sources for the 1.5-4 μm spectral range in the near- to mid-IR based on ultrafast synchronously-pumped optical parametric oscillator (SPOPO) concept in the picosecond time-scale. This line of research was motivated by the high average powers and increased peak pulse intensities that would be potentially available from such SPOPOs, thus more readily facilitating the successful operation of secondary OPOs for wavelength generation beyond 5 μm . We exploited the latest generation of QPM nonlinear materials in combination with high-average-power mode-locked picosecond Yb fiber lasers at 1064 nm, to provide practical, high-power and portable device architectures.

To achieve this goal, we first developed a high-power picosecond SPOPO pumped by a mode-locked Yb fiber laser at 1064 nm, providing 11.7 W of total average power in the near- to mid-infrared at 73% extraction efficiency [5]. The SPOPO, based on a 50 mm MgO:PPLN crystal, was pumped by 20.8 ps pulses at 81.1 MHz, and could simultaneously deliver 7.1 W of signal at 1.56 μm and 4.6 W of idler at 3.33 μm for 16 W of pump power. The SPOPO had a threshold of 740 mW, with maximum signal power of 7.4 W at 1.47 μm and idler power of 4.9 W at 3.08 μm at slope efficiencies of 51% and 31%, respectively. Wavelength coverage across 1.43-1.63 μm (signal) and 4.16-3.06 μm (idler) was obtained, with total power of ~ 11 W and extraction efficiency of $\sim 68\%$, with pump depletion of $\sim 78\%$ maintained over most of the tuning range. The signal and idler output were characterized by single-mode spatial profile and peak-to-peak power

stability of $\pm 1.8\%$ and $\pm 2.9\%$ over 1 hour at the highest power, respectively. A signal pulse duration of 17.3 ps with a clean single-peak spectrum resulted in a time-bandwidth product of ~ 1.72 , more than four times below the input Yb fiber pump pulses. Detailed discussion of the design parameters and performance characteristics of this SPOPO has already been presented in interim Progress Report 4.

4. Multiwatt, fiber-laser-pumped, near- to mid-infrared, synchronously-pumped picosecond optical parametric oscillator based on MgO:sPPLT

Following the successful development of the first prototype, we extended this approach to a SPOPO based on MgO:sPPLT as the nonlinear gain material, using the same mode-locked picosecond Yb fiber laser at 1064 nm as the pump source [6]. Using a 30-mm-long crystal, we achieved a tunable range over 1531-1642 nm (111 nm) in the near-infrared signal and 3022-3488 nm (466 nm) in the mid-infrared idler, providing a total tuning range of 577 nm. Careful optimization of output coupling resulted in a signal output power as high as 4.3 W at 1593 nm and a mid-infrared idler power of 2 W at 3204 nm for 13.4 W of pump power at a total extraction efficiency of 47%. The SPOPO could be operated near room temperature, down to 30 °C, and exhibited passive peak-to-peak power stability better than 8.6 % at 1568 nm (signal) and 8.2% at 3310 nm (idler) over 13 hours at full power. The output signal pulses were measured to have durations of 17.5 ps, with a FWHM spectral bandwidth of 1.4 nm centered at 1568 nm.

4. 1. Experimental setup

The schematic of the experimental setup is similar to that used in our earlier work [5]. The pump source is a mode-locked Yb-fiber laser (Fianium FP1060-20) providing up to 20 W of output power in 20 ps pulses at a repetition rate of 81.1 MHz. The laser has a double-peak spectrum and operates at 1064 nm with an FWHM bandwidth of 1.38 nm, which is well below the spectral acceptance bandwidth of >2.5 nm for optical parametric generation in the 30-mm MgO:sPPLT crystal used in our experiment. The crystal is 1-mm-thick with six gratings, ranging in period from $\Lambda=29.15$ μm to 30.65 μm , and is housed in an oven whose temperature can be controlled in steps of ± 0.1 °C. The SPOPO is configured in a four-mirror standing-wave cavity comprising two plano-concave mirrors ($r=150$ mm), one plane mirror and an output coupler. All mirrors have high reflectivity ($R>99\%$) over 1.3–1.9 μm and high transmission ($T>90\%$) over 2.2–4 μm , ensuring singly resonant oscillation. The pump beam is focused to a beam waist radius of 68 μm ($\xi\sim 0.5$) at the center of the nonlinear crystal, with a corresponding signal beam waist radius of 83 μm ($b_p\sim b_s$). A dichroic mirror separates the generated idler from the pump, while the signal

power is extracted from the output coupler (OC). The total round-trip optical length of the cavity is ~ 370 cm, ensuring synchronization with the pump laser.

4. 2. Wavelength tuning and extracted power

In order to characterize the SPOPO, we initially performed temperature tuning for two grating periods, $\Lambda=30.65$ and 30.15 μm , by varying the crystal temperature. The signal wavelength was measured using a near-infrared spectrum analyzer and the corresponding idler wavelength was calculated from energy conservation. Figure 6 shows the measured output power across the tuning range of the picosecond MgO:sPPLT SPOPO with a maximum pump power of 13.6 W at the input to the crystal. In order to achieve signal power extraction across the full SPOPO tuning range, we deployed two OCs available in our laboratory. Figure 6(a) shows the extracted signal power with OC₁ (T $\sim 10\%$ over 1531 - 1568 nm) and OC₂ (T ~ 25 - 70% over 1572 - 1642 nm), together with the transmission of the OCs used to access the wide tuning range of the SPOPO. Using the two grating periods, $\Lambda=30.65$ and 30.15 μm , and by changing the crystal temperature from 30 $^{\circ}\text{C}$ to 200 $^{\circ}\text{C}$, we were able to tune the SPOPO over 1531 - 1642 nm in the signal and 3022 - 3488 nm in the idler wavelength range, resulting in a total (signal plus idler) tuning of 577 nm. We obtained good agreement between the theoretical temperature tuning curves calculated from the relevant Sellmeier equations [4] and the experimentally measured data. As shown in Fig. 6(a), the signal power extracted by deploying OC₁ is ~ 3 W and is constant over the range 1531 - 1568 nm, implying that the output power is limited by the transmission of OC₁. With OC₂, the signal power varies from 3.9 W at 1572 nm to 3.2 W at 1642 nm, with maximum power obtained for an output coupling of 27 - 60% . It is interesting to note that the variation in the signal power in this output coupling range is nominal, indicating that the SPOPO is somewhat insensitive to losses. However, an overall comparison of the OC transmission and the extracted signal power shows that the variation of the signal power is not solely governed by the OC. The maximum signal power achieved was 4.2 W for an output coupling of $\sim 47\%$. The corresponding idler power varies from 1.93 W at 3022 nm to 1.88 W at 3488 nm, with a maximum of 2.26 W at 3283 nm, as shown in Fig. 6(b), together with the pump depletion of $>50\%$ recorded over 61% of the idler tuning range.

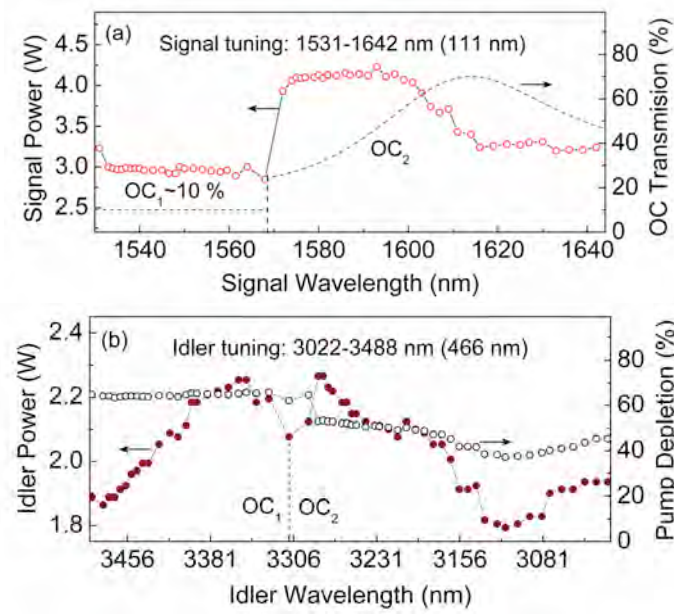


Fig. 6. Variation of (a) signal power together with transmission of different OCs, (b) idler power and pump depletion across the tuning range.

4. 3. Power scaling and optimization

We also optimized the output coupling at a fixed signal wavelength, using $\Lambda=30.65 \mu\text{m}$, by employing different OCs with signal transmission from $T\sim 5\%$ to 88% . The measurements were performed at $T=100^\circ\text{C}$, resulting in a signal wavelength of 1593 nm , corresponding to an idler wavelength of 3204 nm . Figure 7(a) shows the extracted signal power for different OCs at a maximum available input pump power of 13.3 W . Also shown in the inset of Fig. 7(a) is the simultaneously extracted idler power. As evident from the plot, an increase in the OC transmission from $\sim 5\%$ to $\sim 47\%$ resulted in an increase in the signal power from 2.4 W to 4.23 W , without significant compromise in the idler power remaining above 2 W . Further increase in the output coupling to 88% led to the drop in signal power down to 3.32 W , while the corresponding idler power decreased to 1.56 W . This implies an optimum signal output coupling of $\sim 47\%$ for our SPOPO at this wavelength, consistent with the data in Fig. 6(a). For this value of optimum output coupling, the extraction efficiency is 46.9% , and the threshold pump power is recorded to be 2.8 W . Hence, the SPOPO is pumped ~ 4.8 times above threshold. This value is close the theoretical prediction for a Gaussian pump beam, where a maximum down-conversion efficiency of 71% is expected when pumping ~ 6.5 times above threshold [7]. In our SPOPO, the maximum pump depletion is $>50\%$ when pumping ~ 4.8 times threshold. One could adjust the output coupling so that the SPOPO is pumped at ~ 6.5 times above threshold, for the highest pump depletion. However, the focus of our study was the maximization of SPOPO output power and extraction efficiency. Further optimization of output coupling at different wavelengths

across the SPOPO tuning range and for different pumping levels can be performed using interferometric techniques [8,9]. We also investigated power scaling of the SPOPO at the optimum output coupling, for $\Lambda=30.65 \mu\text{m}$, with the results shown in Fig. 7(b). In case of optimum output coupling of $\sim 47\%$, we extracted as much as 4.28 W of signal power at 1593 nm, together with 2 W of mid-IR idler power at 3204 nm, for a pump power of 13.4 W, corresponding to a total power of 6.28 W, representing a total extraction efficiency of 46.9%, consistent with the measurements in Fig. 7(a). The threshold pump power for the optimally output-coupled SPOPO is 2.8 W, and the slope efficiencies of the extracted signal and idler power are 36.4% and 14.5%, respectively.

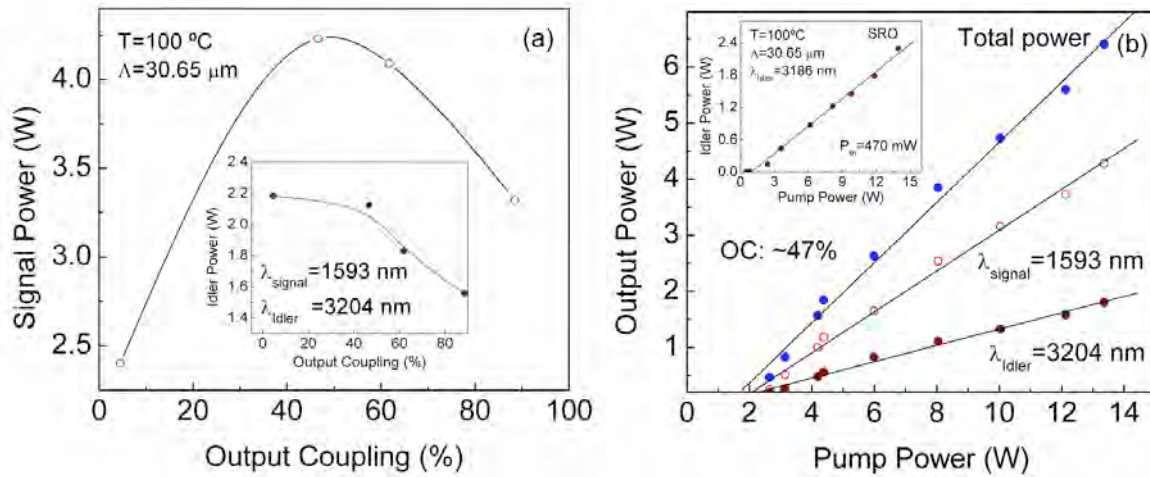


Fig. 7. (a) Variation of signal and (Inset) idler power from the picosecond MgO:sPPLT SPOPO as a function of output coupling for $\Lambda=30.65 \mu\text{m}$. (b) Power scaling of the SPOPO at an optimum output coupling of $\sim 47\%$. Inset: Power scaling in the absence of signal output coupling.

On the other hand, similar measurements for SPOPO with no output coupling resulted in a maximum idler power of 2.3 W at 3186 nm for a pump power of 13.9 W, as shown in the inset of Fig. 7(b), using the same grating period, $\Lambda=30.65 \mu\text{m}$, at $T=100^\circ\text{C}$. Under this condition, the SPOPO threshold pump power was as low as 450 mW. Also to be noted in Fig. 7(b) is the difference in the idler wavelengths when the SPOPO is operating with no output coupling (3186 nm) compared to that in the case of optimum output coupling (3204 nm). The corresponding signal wavelengths in the un-output-coupled and optimally output-coupled SPOPO are 1597 nm and 1593 nm, respectively. This difference in wavelength, despite SPOPO operating at the same temperature, $T=100^\circ\text{C}$ in both configurations, is attributed to the change in crystal temperature due to the small absorption at the signal wavelength due to significantly different intracavity power levels in the two configurations.

We also investigated the performance of the output-coupled SPOPO when operating close to room temperature. Figure 8 shows the variation of the signal, idler and total power as a function of pump power at $T=30\text{ }^{\circ}\text{C}$ for a grating period of $\Lambda=30.15\text{ }\mu\text{m}$, generating signal and idler wavelengths of 1531 nm and 3488 nm, respectively. These measurements were performed with a 10% OC, resulting in the extraction of 3.23 W of signal power at a slope efficiency of 25.8%, together with 1.86 W of mid-IR idler power at a slope efficiency of 15.3%, representing a total power of 5.09 W for a pump power of 13.7 W. The threshold of the SPOPO in this case was 1.87 W, owing to the 10% output coupling.

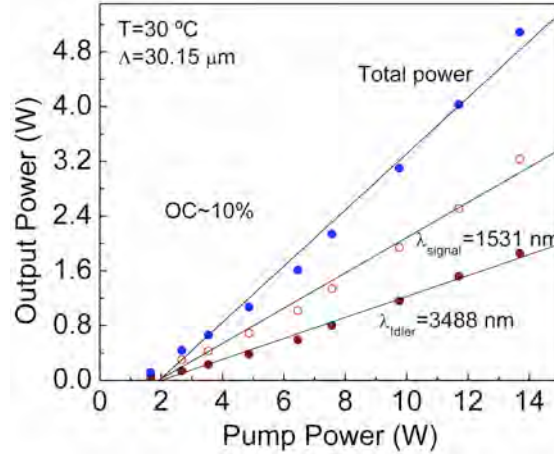


Fig. 8. Power scaling of picosecond MgO:sPPLT SPOPO using the $\Lambda=30.15\text{ }\mu\text{m}$ grating, with an output coupling of $\sim 10\%$.

Further, we compared the SPOPO performance at the same wavelength, while operating at different temperatures, by using two different grating periods. The high temperature operation of the SPOPO with $\Lambda=30.65\text{ }\mu\text{m}$ grating, was compared with the low temperature operation using $\Lambda=30.15\text{ }\mu\text{m}$ grating, generating the same signal and idler wavelengths. For example, at a temperature of $170\text{ }^{\circ}\text{C}$, using a grating period of $\Lambda=30.15\text{ }\mu\text{m}$, signal wavelength of 1572 nm, with a corresponding idler wavelength of 3273 nm, was generated. The extracted signal and idler powers under this condition were 3 W and 2.2 W, respectively, representing a total (signal plus idler) power of 5.2 W for an input pump power of 13.3 W. The same wavelengths can also be generated by using the grating period $\Lambda=30.65\text{ }\mu\text{m}$ and operating the SPOPO at $30\text{ }^{\circ}\text{C}$, close to room temperature. Under this condition, we were able to extract 3.9 W of signal together with 2.2 W of idler, representing a total power of 6.1 W for the same pump power. This confirms that the performance of the SPOPO is significantly better near room temperature than at high temperatures with regard to output power and extraction efficiency.

4. 4. Power stability and temporal characteristics

We recorded the long-term power stability of the picosecond MgO:sPPLT SPOPO close to room temperature, at $T=30\text{ }^{\circ}\text{C}$, using OC₂ at a signal wavelength of 1568 nm, corresponding to mid-IR

idler wavelength of 3310 nm, simultaneously at full output power. The result is shown in Fig. 9(a), where a peak-to-peak power stability of 8.6% for the signal and 8.2% for the idler over a period of 13 hours is obtained under free-running conditions. The long-term drift in the signal and idler power is due to the thermal and mechanical fluctuations effecting the cavity length and alignment of the SPOPO. However, realignment of the cavity at the end of the stability measurement resulted in the operation of the SPOPO with original performance. Hence, isolation from thermal and mechanical fluctuations will further improve the stability of the SPOPO [5].

Finally, we performed spectral and temporal characterization of the signal pulses from the picosecond MgO:sPPLT SPOPO. Figure 9(b) shows the typical interferometric autocorrelation, with signal pulse duration of $\Delta\tau \sim 17.5$ ps. The corresponding signal spectrum with a FWHM bandwidth of 1.4 nm, centered at 1568 nm, is shown in the inset of Fig. 9(b). These measurements correspond to a time-bandwidth product of $\Delta\nu\Delta\tau \sim 3$ for the signal pulses, which can be further improved by implementing dispersion compensation.

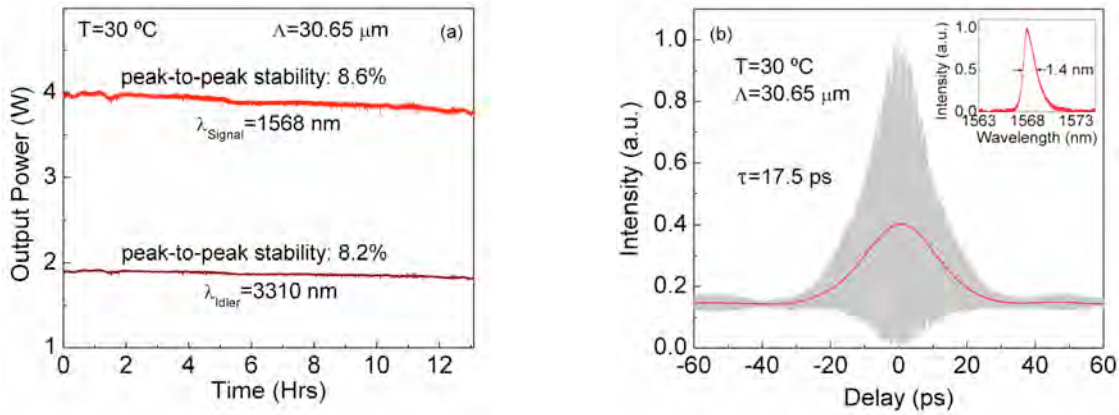


Fig. 9. (a) Long-term power stability of the signal and idler from the picosecond MgO:sPPLT SPOPO at $T=30$ °C. (b) Interferometric autocorrelation of the output signal pulses from the picosecond MgO:sPPLT SPOPO, and (inset) corresponding signal spectrum centered at 1568 nm.

The successful development of the high-power cw OPOs and high-average-power picosecond SPOPOs developed during the project, as described in sections 1-4 above, has led to the realization of high-power sources of tunable coherent radiation for the near- to mid-infrared using highly practical, compact, and portable architectures based on fiber laser technology, and capable of providing multiwatt optical power levels across the $1.5\text{--}4\text{ }\mu\text{m}$ spectral range with high passive stability and potential for power scaling. The availability of these sources has provided an important step, paving the way for their exploitation as primary pumps for secondary OPOs to achieve wavelength generation beyond $5\text{ }\mu\text{m}$. With our most recent procurement of mid-IR nonlinear crystals of OP-GaAs and ZnGeP₂ of high optical quality and low transmission loss, this work is now in progress. We, thus, expect to achieve successful operation of

secondary OPOs based on OP-GaAs and ZnGeP₂ in the coming months, providing cw and high-repetition-rate picosecond pulsed radiation beyond 5 μm in the mid-IR spectral regions.

5. High-repetition-rate, picosecond optical parametric generator beyond 5 μm

In addition to the cascaded two-step OPO concept, a complementary approach involving direct single-step pumping was also investigated during the project. While the exploitation of oxide-based birefringent materials such MgO:PPLN and MgO:sPPLT permits access to spectral regions up to $\sim 5 \mu\text{m}$ through parametric down-conversion, the onset of multiphonon absorption sets a practical upper limit of $\sim 4\text{--}4.5 \mu\text{m}$ for wavelength generation in such materials. The use of chalcogenide nonlinear crystals with transparency in the mid-IR, such as CdSe and AgGaSe₂, can provide a solution for coherent light generation at longer wavelengths, but the low bandgap energy in these materials precludes pumping near $\sim 1 \mu\text{m}$ due to two-photon absorption (TPA), thus preventing the deployment of practical solid-state Nd-based pump lasers. Other chalcogenide crystals with larger bandgap, such as AgGaS₂, may be pumped near $\sim 1 \mu\text{m}$ without the onset of TPA, but the poor thermomechanical properties including low thermal conductivity, anisotropy of thermal expansion, and low damage threshold prevent practical device operation. As such, exploitation of many chalcogenide materials requires long-wavelength laser pump sources with limited availability near $\sim 2 \mu\text{m}$, or the deployment of cascaded pumping schemes with the associated complexities. In this context, the newly discovered nonlinear material, CdSiP₂ (CSP) offers unique linear and nonlinear properties for parametric down-conversion into the mid-IR [10]. It is a negative uniaxial chalcopyrite compound with a transparency above $\sim 6.5 \mu\text{m}$, which possesses noncritical phase-matching (NCPM) capability with a maximum effective nonlinear coefficient as high as $d_{\text{eff}}=d_{36}=84.5 \text{ pm/V}$ [11]. Importantly, CSP has a band-gap well below $1 \mu\text{m}$, which permits pumping at 1064 nm, and under type I ($e \rightarrow oo$) parametric generation with NCPM can provide an idler wavelength beyond $5 \mu\text{m}$ in the mid-IR range. In this project, we successfully exploited the unique properties of this new nonlinear material to provide tunable coherent radiation at wavelengths in the $6\text{--}6.5 \mu\text{m}$ region using optical parametric generation and oscillation pumped at 1064 nm.

By using an amplified mode-locked picosecond Nd:YVO₄ laser at 1064 nm and 100 kHz repetition rate as the pump source and single-pass optical parametric generation in 8-mm-long crystal cut for type I ($e \rightarrow oo$) noncritical phase-matching, we achieved an average idler power of 154 mW at 6204 nm together with 1.16 W of signal at 1.282 μm for 6.1 W of pump at photon conversion efficiencies of 15% and 25%, respectively [12]. The device generated signal pulse

durations of 6.36 ps for 9 ps pump pulses, with both signal and idler beams in near-Gaussian spatial profile. Detailed discussion of the design parameters and performance characteristics of this device has already been presented in interim Progress Report 4.

6. High-energy, high-repetition-rate, synchronously-pumped picosecond optical parametric oscillator beyond 5 μm

Following the breakthrough demonstration of mid-IR wavelength generation beyond 5 μm using OPG in CSP, we were able to successfully develop a compact, high-energy, high-repetition-rate picosecond SPOPO pumped by a Nd:YAG/Nd:YVO₄ oscillator/amplifier system at 1064 nm [13]. The pump source for this SPOPO delivered 1- μs -long, macro-pulses constituting 10 ps micro-pulses at 450 MHz repetition rate. Using a compact (~ 30 cm) cavity and improved, high quality, 12.1-mm-long CSP crystal, we obtained idler wavelength tuning over 486 nm across 6091-6577 nm in the mid-IR. We generated an idler macro-pulse energy as much as 1.5 mJ at 6275 nm at a photon conversion efficiency of 29.5% for an input macro-pulse energy of 30 mJ, with >1.2 mJ available over more than 68% of the tuning range. Both the signal and idler beams were recorded to have good beam quality with TEM₀₀ spatial profile, and the extracted signal pulses were measured to have durations of 10.6 ps. From the experimentally measured transmission data at 1064 nm, we also estimated a two photon absorption coefficient of $\beta=2.4$ cm/GW and corresponding energy band gap of $E_g=2.08$ eV for the CSP crystal. Detailed discussion of the design parameters and performance characteristics of this SPOPO has already been presented in interim Progress Report 4.

7. High-energy, picosecond optical parametric generator beyond 5 μm

Most recently, we have successfully generated picosecond pulses beyond 5 μm at record energy using single-pass optical parametric generation of in CSP pumped by a cavity-dumped, mode-locked Nd:YAG laser at 1064 nm [14]. The device can provide mid-IR (idler) tuning over 6153-6732 nm and in the near-IR (signal) tuning across 1264-1286 nm. It can deliver ~ 600 μJ of output energy over the entire signal tuning range in 24 ps pulses and >25 μJ of long-wavelength mid-IR pulse energy over 55 % of the idler tuning range.

7. 1. Experimental setup

The pump source is a laboratory designed, cavity-dumped, passively mode-locked, diode-pumped Nd:YAG oscillator, as shown in Fig. 10. The oscillator generates single pulses with energies up to 25 μJ in 20 ps duration, at a repetition rate of 5 Hz, limited by the amplifier flash-

lamp power supply. The pump pulse energy is further boosted by a flash-lamp-pumped Nd:YAG amplifier to 2.5 mJ. The pump laser operates at a central wavelength of 1064 nm and the output beam from pump laser has a diameter of 2 mm with a beam quality factor of $M^2 \sim 1.8$.

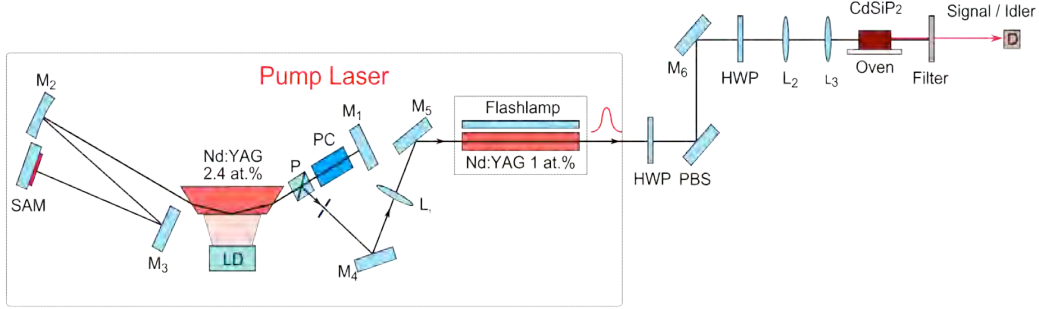


Fig. 10. Experimental setup of the high-energy optical parametric generator.

The nonlinear crystal is a 12.1-mm-long, 4-mm-wide (along the c -axis), 5-mm-thick CSP sample grown from stoichiometric melt by the horizontal gradient freeze technique. It is cut at $\theta=90^\circ$, $\phi=45^\circ$ for type-I ($e \rightarrow oo$) interaction under noncritical phase-matching and housed in an oven whose temperature can be varied from room temperature to 200°C in steps of $\pm 0.1^\circ\text{C}$. Both crystal faces are antireflection-coated with a single layer sapphire coating, providing high transmission ($T > 98.7\%$) for the pump and signal over 1064-1300 nm and $T > 76\%$ for the idler over 6000-6500 nm. The pump beam is collimated to a beam radius of $700 \mu\text{m}$ at the center of the nonlinear crystal. The beam waist is optimized to use the maximum available pump pulse energy and yet to remain below the damage threshold of the nonlinear crystal. The generated signal is separated from the residual pump using a near-infrared filter, while a germanium filter is used to extract the mid-IR idler.

7. 2. Idler wavelength tuning and spectral characteristics

In order to characterize the OPG with regard to wavelength tunability, we initially performed temperature tuning using a thermocouple connected to the crystal holder, to accurately monitor the temperature of the nonlinear crystal. The variation of the oven temperature from 25 to 200°C corresponded to a thermocouple temperature variation from 25 to 174°C . Hence, by changing the temperature of the CSP crystal from 25 to 174°C we were able to tune the OPG over 1264-1286 nm in the near-infrared signal along with a mid-IR idler tunability over 6153-6732 nm, corresponding to a total signal plus idler tuning of 601 nm. Figure 11(a) shows the amount of signal and idler energy extracted over the entire tuning range. As evident from Fig. 11(a), for a fixed pump energy of 2.1 mJ at the input to the CSP crystal, the signal energy remains almost constant at $\sim 600 \mu\text{J}$ over the entire tuning range, while the idler energy varies from the $33 \mu\text{J}$ at 6153 nm to $20 \mu\text{J}$ at 6731 nm, remaining $> 25 \mu\text{J}$

over 55% of the tuning range. The threshold of the OPG is recoded to be $\sim 100 \mu\text{J}$. Additionally, we have recorded the idler spectrum at the same thermocouple temperature of 91°C using a grating spectrometer. The measured idler spectrum center at 6404 nm is shown in Fig. 11(b) and has an FWHM bandwidth of 140 nm .

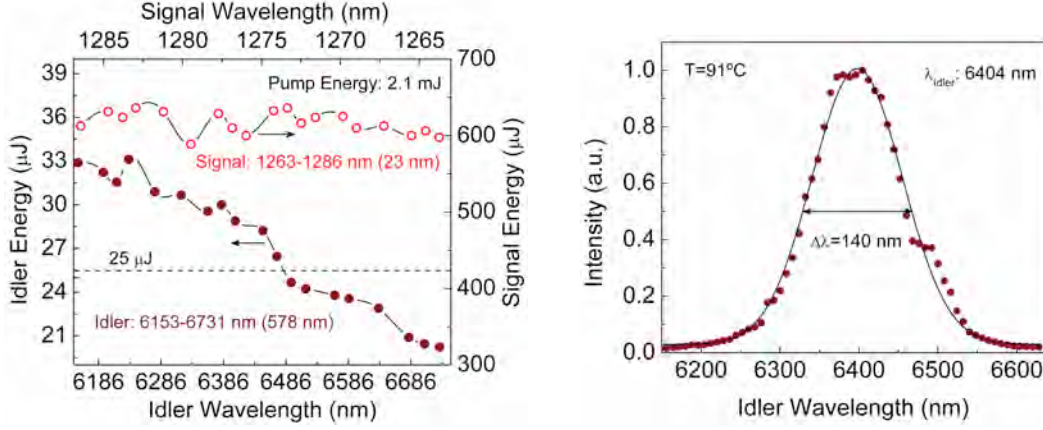


Fig. 11. (a) Signal and idler energy across the tuning range, (b) idler spectrum centered at 6404 nm .

7. 3. Signal temporal and spectral characteristics

Further, we performed spectral and temporal characterization of the signal pulses generated from the OPG. Figure 12(a) shows a typical autocorrelation profile measured at an oven temperature of 105°C corresponding to a thermocouple temperature of 91°C . This corresponds to a signal wavelength of 1276 nm . The FWHM of the trace is 34 ps , resulting in signal pulse duration of 24 ps , assuming a Gaussian pulse shape. This value of the pulse duration was confirmed by repeating the measurement several times. Similar pulse duration is expected across the tuning range. The corresponding signal spectrum measured using a near-infrared spectrometer is centered at 1276 nm with a FWHM bandwidth of 10.4 nm , as shown in Fig. 12(b).

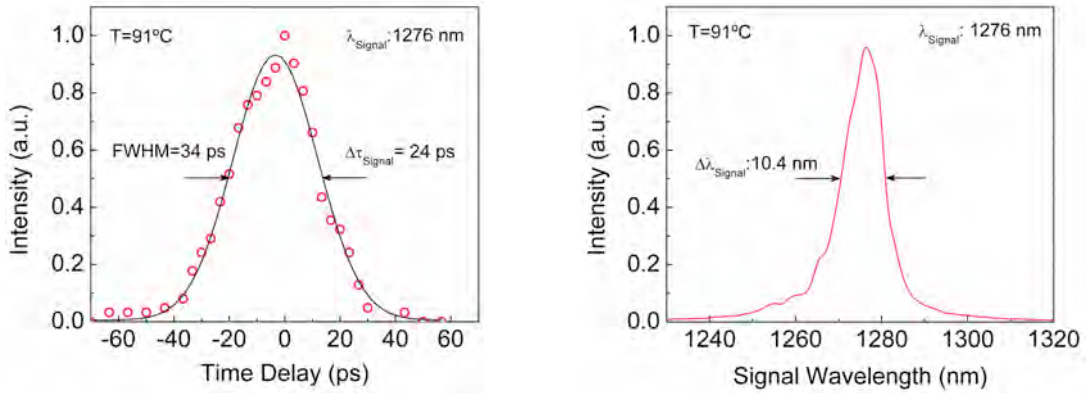


Fig. 12. (a) Typical autocorrelation, (b) spectrum of the OPG signal pulses and at 1276 nm .

8. Summary and Conclusions

This project has led to important new advances in coherent near- to mid-infrared light sources on several fronts. By exploiting nonlinear frequency down-conversion techniques based on optical parametric generation and oscillation, we have developed several novel optical sources in different time-scales and operating regimes with extended tuning throughout the 1.4-4.2 μm spectral range in the near- to mid-IR and 6-6.8 μm in the deep mid-infrared. The developed devices exploit the new generation of QPM and mid-IR nonlinear crystals of MgO:PPLN, MgO:sPPLT, and CdSiP₂ together with fiber and solid-state laser technology at 1064 nm as the pump source, resulting in compact, practical, and portable device architectures for many real applications. We have achieved record optical powers of >17 W in cw operation and average powers up to 12 W in the ultrafast picosecond regime at 80 MHz high-repetition-rate, covering the spectral range of 1.4-4.2 μm . In the deep mid-IR, we have achieved record pulse energies as much as 1.5 mJ at repetition rates as high as 450 MHz, with spectral coverage across 6-6.8 μm . The developed systems have provided a viable new class of tunable coherent light sources for previously inaccessible spectral regions in the near- to mid-infrared offering record optical powers and pulse energies. With the exploitation of mid-IR optical crystals of ZnGeP₂ and OP-GaAs of high optical quality in cascaded pumping schemes using the near- to mid-IR parametric sources developed during this project, further wavelength extension into the 5-12 μm spectral range will be feasible. The exploitation of newly emerging Tm fiber laser technology near 2 μm also offers great potential for direct generation of tunable coherent radiation at unprecedented optical powers in the 5-12 μm spectral range using one-step parametric generation in the nonlinear crystals of ZnGeP₂, CdSiP₂, and OP-GaAs. These exciting new avenues for further research and development are currently underway.

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13. M. Ebrahim-Zadeh, "Optical Parametric Oscillators: After nearly 50 years," Alexander von Humboldt Forum, Science and Society in Modern Europe, Vilnius, Lithuania, September 2010
14. M. Ebrahim-Zadeh, G. K. Samanta, S. Chaitanya Kumar, "Advances in continuous-wave optical parametric oscillators," International Conference on Fiber Optics and Photonics, Photonics 2010, Guwahati, India, December 2010
15. M. Ebrahim-Zadeh, "Optical parametric oscillators," Conference on Lasers and Electro-Optics (CLEO)-Europe, Munich, Germany, May 2011 - Short Course
16. M. Ebrahim-Zadeh, "Optical parametric oscillators spanning the ultraviolet to mid-infrared," Conference on Lasers and Electro-Optics (CLEO)-Europe, Munich, Germany, May 2011
17. M. Ebrahim-Zadeh, "Optical parametric oscillators: New advances," 10th Euro-American Workshop on Information Optics (WIO'2011), Benicassim, Spain, June 2011
18. M. Ebrahim-Zadeh, "Advances in Optical Parametric Oscillators", 20th International Laser Physics Conference, LPHYS'11, Sarajevo, Bosnia-Herzegovina, July 2011.

Invited Seminars

1. M. Ebrahim-Zadeh, "Nonlinear optics: A powerful tool for manipulation of laser light", Helsinki University of Technology, Finland, November 2009
2. M. Ebrahim-Zadeh, "Laser light conversion and manipulation with nonlinear optics," Institute of Applied Physics, University of Bonn, Germany, June 2010.
3. M. Ebrahim-Zadeh, "The laser: From Maiman to modern times," LaserFest Lecture, Institute of Photonic Sciences, Barcelona, Spain, September 2010.
4. M. Ebrahim-Zadeh, "Acquiescent light," Max-Planck Institute for Quantum Optics, Garching, Germany, October 2010.

International Prize

M. Ebrahim-Zadeh, Berthold Leibinger Innovation Prize, Germany, July 2010